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TECHNICAL REPORT

AMRA TR 63-03

CONTRIBUTION OF THERMAL DIFFUSE SCATTERING TO INTEGRATED BRAGG REFLECTIONS FROM PERFECT CRYSTALS

by

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JANUARY 1963 WATERTOWN 72, MASS.

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X-ray scattering

Phonons

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Technical Report AMRA TR 63-03

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ABSTRACT

It is shown that the contribution of thermal diffuse scattering to integrated Bragg intensities from perfect crystals can be much greater than previously expected. Measurements on the (555) reflection of silicon are presented showing a thermal scattering contribution of about 70% of the true Bragg intensity. The special aspect of the effect in perfect crystals is discussed and methods of reducing it or correcting for it are included.

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INTRODUCTION

Thermal diffuse scattering (TDS) of X-rays or neutrons generally makes up an important part of what is called the "background" in a diffraction pattern. This diffuse scattering is not independent of angle, but has peaks at the same places where Bragg reflections occur. As shown by Nilsson[1] and by Chipman and Paskin[2] these peaks will lead to errors in measured integrated Bragg intensities if a straight line background under the reflection is erroneously assumed. For example, an error of 15% for the (511) reflection of lead powder has been reported.[2]

It is the purpose of this note to point out that for crystals of high perfection, this effect can be many times larger. Starting from an imperfect crystal, as the perfection is increased, extinction increases, and Bragg intensities decrease. However, the TDS intensity function is very weak, so that it is scarcely affected by extinction and its value, even in a perfect crystal, is given by a mosaic crystal formula. Therefore, as perfection is increased, the TDS peak becomes a relatively larger fraction of the Bragg peak, resulting in an enhancement of the effect. This enhancement can become as large as the ratio of integrated intensities calculated from the mosaic and perfect crystal formulas.[3]

In practice, the occurrence or at least the observation of this effect depends markedly on the experimental geometry. The TDS peaks are very broad functions on an angular scale, and the TDS contribution to a Bragg peak varies almost linearly[2] with the total range of integration, i.e., the total length of the background line drawn under the peak. Therefore, if a perfect crystal is measured in a double crystal spectrometer in the parallel position, the TDS contribution will seem to be very small since the total width of the scan is typically only a few tens of seconds of arc. Of course if the scan were actually carried out over a range of several degrees, the TDS peak would be observed and the effect discussed above would be obtained. However, with such a narrow peak there is little temptation to do so.

In contrast to the above case, Fig. 1 shows the (555) reflection of perfect crystal silicon obtained by rocking the crystal through the entire reflecting range in a monochromatic primary beam of about one degree divergence. Here the Bragg reflection is spread out instrumentally over about two degrees in 2θ, and the TDS becomes quite apparent. It will be shown below that the true Bragg peak is approximately that area above the dotted line, the rest being contributed by TDS.

The intensity scale of Fig. 1 was determined in absolute units by a direct measurement of the primary beam using absorbers. Several methods are therefore available by which we can determine the relative amounts of TDS and Bragg reflection in Fig. 1. First, we calculated the TDS in the vicinity of the reflection by making use of the measured value of the Debye-Waller factor of silicon.[4] A brief discussion of the calculation and the assumptions made is given in the Appendix. The results of the calculation are shown as the solid points in Fig. 1.

Second we have compared the measured integrated intensity of the reflection in Fig. 1, with an experimental value which we have measured using a double crystal spectrometer, and with the theoretical (Darwin-Prins) value. The comparison is made by measuring the area above a straight line which is allowed to range between the solid and the dotted lines of Fig. 1, while remaining parallel to both. This area is plotted against the length of the corresponding background line, as shown in Fig. 2. This figure shows that as the range of integration is narrowed, the intensity approaches the experimental double crystal or theoretical values.

Third, we have used a theoretical expression which gives the ratio of the TDS area to the Bragg peak area for a powder sample.[2] This expression should not, of course, be expected to give the exact correction for a single crystal, but it is probably not greatly in error. Since the expression is derived for the TDS correction to ideally imperfect crystal Bragg peaks, we increase it by the ratio of the intensities calculated from the mosaic and perfect crystal formulas so that it will apply to a perfect crystal. This factor for the (555) reflection of silicon and MoKα radiation is about 7. This expression gives the fraction of the area which is TDS as a function of the width of integration. In Fig. 2, we have drawn it to intercept the intensity axis at the double crystal spectrometer value, from which it matches the initial data points quite well.

These three different approaches all lead to the same conclusion, that the area above the straight line background in Fig. 1 is some 70% too large, and that this extra scattering is a result of TDS. This enhancement, which occured because the crystal was perfect, has thus changed what might have been thought of as an almost negligible correction into a very important one. For the (111) reflection, the factor is about 27, which increases a mosaic crystal TDS correction of 0.15% for our geometry up to about 4%, a value which we have also verified experimentally.

In conclusion, under certain conditions which are not particularly unusual, the TDS correction can be very large. The geometry of the diffraction experiment is quite important in determining how large the contribution of TDS will be, and by giving some thought to this, the contribution may be substantially reduced for a given measurement. In any case, the correction will always become larger when the divergences in the X-ray system are increased or when the perfection of the sample becomes greater.

APPENDIX

In the vicinity of a Bragg reflection, the optic mode scattering is negligible compared with the acoustic mode. Hence only the intensity resulting from the acoustic mode was calculated, by assuming that all elastic waves have the same velocity, and obtaining this velocity from the experimental value of the Debye-Waller factor (Batterman & Chipman, 1962). A curve of TDS intensity versus angle was calculated from this intensity function for the particular geometrical arrangement of the apparatus using a numerical method of averaging over the receiving volume element. All other sources of background scattering near the peak (Compton scattering, etc.) were assumed to be independent of angle, so a constant was added to the calculated TDS to make it match the curve of Fig. 1 at some one angle. Both the TDS and the intensity scale of Fig. 1 had to be known in the same intensity units to make this possible. This was done by measuring the total power in the direct beam using absorbers.[5]

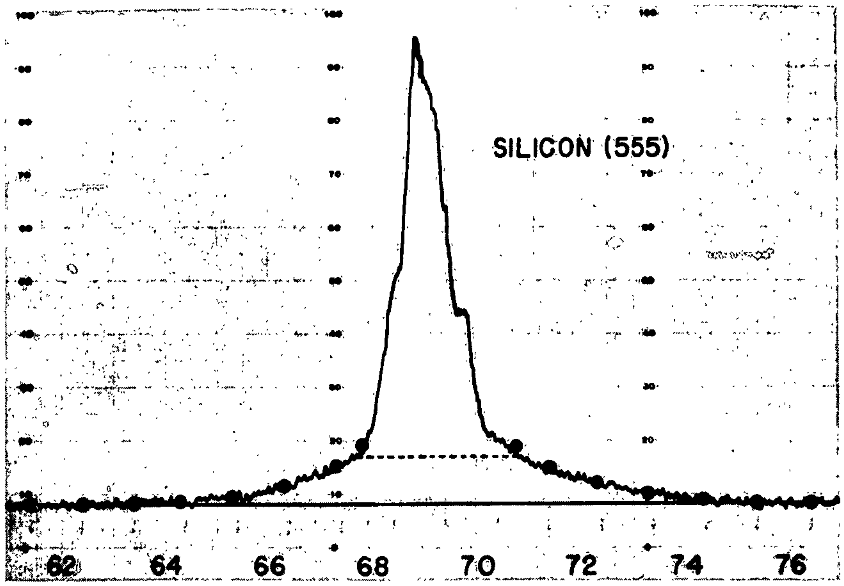

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Figure 1. The (555) reflection of a silicon single crystal measured using a monochromatic primary beam having a divergence of about one degree. The crystal was rocked through the entire reflecting range with a wide open counter. Intensity is plotted versus 2θ. Filled circles are the calculated thermal diffuse scattering.

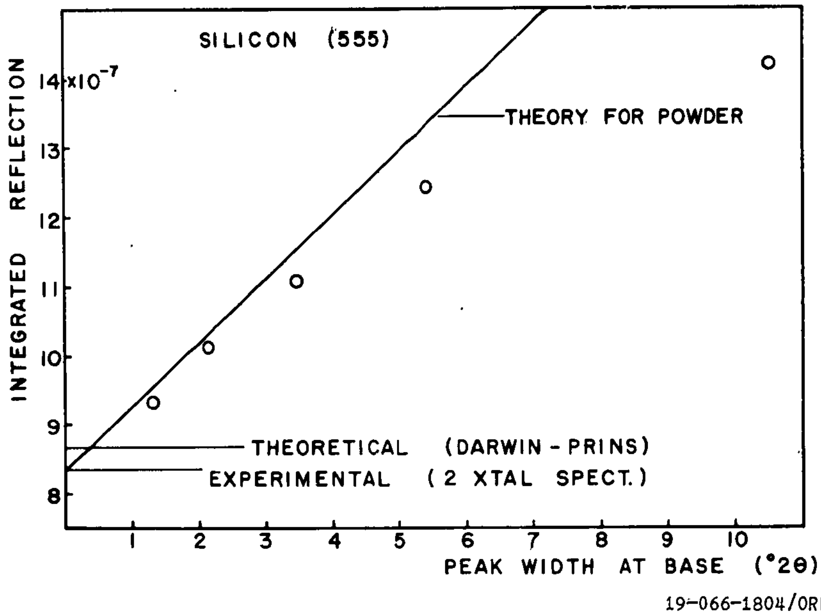

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Figure 2. The integrated intensity from Figure 1 measured above a horizontal line lying between the solid and dotted lines of Figure 1, versus the length of the horizontal line (open circles). Also shown are the theoretical and experimental values for the integrated intensity of the (555) reflection of silicon, and the Chipman & Paskin[2] powder theory.

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